

# Excitation of the isomeric $^{229m}\text{Th}$ nuclear state via an electronic bridge process in $^{229}\text{Th}^+$

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We consider the excitation of the nuclear transition  $^{229g}\text{Th} - ^{229m}\text{Th}$  near 7.6 eV in singly ionized thorium via an electronic bridge process. The process relies on the excitation of the electron shell by two laser photons whose sum frequency is equal to the nuclear transition frequency. This scheme allows to determine the nuclear transition frequency with high accuracy. Based on calculations of the electronic level structure of  $\text{Th}^+$  which combine the configuration-interaction method and many-body perturbation theory, we estimate that a nuclear excitation rate in the range of  $10\text{ s}^{-1}$  can be obtained using conventional laser sources.

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The nuclear transition between the low-energy isomeric state and the ground state in the  $^{229}\text{Th}$  nucleus is very interesting due to the possibility to build a very precise nuclear clock and its high sensitivity to a hypothetical temporal variation of the fundamental constants [1, 2]. The value of the transition frequency is known from  $\gamma$ -spectroscopy [3–5] with an uncertainty that is many orders of magnitude higher than the presumed linewidth. The presently most precise value of the transition energy is 7.6(5) eV [5], placing the transition in the vacuum-ultraviolet spectral range. Further investigations are therefore needed in order to allow a direct optical excitation and detection of the line and to gradually increase the spectroscopic resolution to the level adequate for an optical clock [1]. Since the energy of this nuclear transition is in the same range as resonances of the outer-shell electrons in thorium ions and chemical compounds of Th, the electronic environment may have a significant influence on the nuclear transition rate [6]. In this work we suggest a new experiment to study laser excitation of the nuclear  $^{229g}\text{Th} - ^{229m}\text{Th}$  transition, making use of the dense electronic level structure of  $\text{Th}^+$ .

In a recent article [7] we have studied the effect of atomic electrons on the nuclear transition from the isomeric  $^{229m}\text{Th}$  state to the ground  $^{229g}\text{Th}$  state in  $^{229}\text{Th}^+$  due to an electronic bridge (EB) process. Here we consider the process of the nuclear  $g \rightarrow m$  excitation via an EB process. Atomic units ( $\hbar = |e| = m_e = 1$ ) are used unless noted otherwise.

**Experimental scheme.** The EB process considered in this work can be represented by the Feynman diagram in Fig. 1. In the following we assume a resonant character of this EB process. For this reason we take into account only one Feynman diagram given by Fig. 1 which mainly contributes to the probability of the process. Other diagrams which can be obtained from this one by permutations of the photon lines will be neglected. We sup-

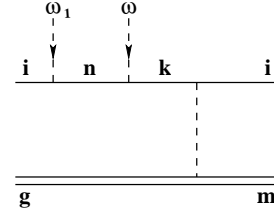


FIG. 1: Feynman diagram of the considered two-photon electronic bridge process. The single and double solid lines relate to the electronic and the nuclear states, respectively. The dashed lines are the photon lines.

pose that the initial state  $i$  is the electronic ground state and the initial and final electronic states are the same. Further we assume a two-photon excitation of high-lying states  $k$ . For the first excitation at  $\omega_1$ , it seems convenient to use a continuous laser tuned to the transition from the  $\text{Th}^+$  ground state ( $6d^27s$ )  $J = 3/2$  to the ( $6d7s7p$ )  $J = 5/2$  state at  $24874\text{ cm}^{-1}$  [8]. This electric dipole (E1) transition is known as the strongest emission line in the  $\text{Th}^+$  spectrum and it can be excited efficiently in collisionally cooled ion clouds in a radiofrequency trap [9]. As a result, only the ( $6d7s7p$ )  $J = 5/2$  state needs to be accounted for in the sum over intermediate states  $n$  in Fig. 1. We also note that the monitoring of the fluorescence resulting from excitation of the ( $6d^27s$ )  $J = 3/2 \rightarrow (6d7s7p)$   $J = 5/2$  transition allows us to detect the excitation of  $^{229}\text{Th}^+$  ions from the nuclear ground state to the isomeric state, making use of the differences between the hyperfine structure characteristics of electronic transitions in  $^{229m}\text{Th}^+$  and in  $^{229g}\text{Th}^+$  [1].

The second excitation step with frequency  $\omega$  gives rise to the excitation of higher-lying states  $k$ . Their decay to the ground state is accompanied by the nuclear  $g \rightarrow m$  transition and leads to the appearance of isomeric  $\text{Th}^+$

ions in the electronic ground state. For this excitation scheme, energy conservation implies that  $\omega_1 + \omega = \omega_N$ , where  $\omega_1$  and  $\omega$  are the frequencies of the incident photons, and  $\omega_N = E_m - E_g$  is the nuclear transition frequency as determined by the difference between the isomeric nuclear energy  $E_m$  and the ground-state energy  $E_g$ .

This excitation scheme offers the prospect to determine the nuclear transition frequency  $\omega_N$  with the accuracy afforded by high-resolution laser spectroscopy. In an experiment based on this scheme, one would use a widely tunable laser source to produce narrow-bandwidth radiation with variable frequency  $\omega$ . If the laser frequency is scanned over the nuclear resonance, the probability of excitation to the isomeric state exhibits a resonance peak. In an ion-trap experiment with collisionally cooled  $^{229}\text{Th}^+$  ions, the width of the resonance is determined essentially by Doppler broadening and by the combined linewidth of the employed laser sources. If one assumes saturated excitation to the state  $n$  and a fixed detuning of  $\omega$  relative to the electronic transitions  $n \rightarrow k$ , the probability of the EB process shown in Fig. 1 is proportional to the spectral intensity of the laser field at  $\omega$ .

Based on the result of Ref. [5] we assume that the most interesting range of the electronic excitations is around  $7.6 \text{ eV} \approx 61300 \text{ cm}^{-1}$ . Following this assumption, we expect that in the sum over the intermediate states  $k$  the atomic energy levels lying between  $60\,000$  and  $64\,000 \text{ cm}^{-1}$  will give the predominant contribution to the probability of the EB process. Unfortunately, these energy levels are not yet identified experimentally. Therefore all the following results are based on *ab initio* calculations. As we noted in [7], the achieved accuracy of calculations of the high-lying states of  $\text{Th}^+$  is at the level of several percent. This is not sufficient to reliably predict the resonance enhancement occurring in the EB process. Hence, the experimental identification of the energy levels should be considered as the next step towards realizing the method considered here.

Because at present very accurate calculations are not needed we will make one more assumption simplifying the calculations. We assume that only one intermediate state  $n$ , the state at  $24874 \text{ cm}^{-1}$ , contributes to the probability of the EB process and that 100% population of this state can be achieved. As a result, the frequency  $\omega_1$  is assumed to be fixed at  $24874 \text{ cm}^{-1}$  and the process which we discuss in the following can be described by the diagram represented by Fig. 2, where the state at  $24874 \text{ cm}^{-1}$  is denoted as  $t$  and is considered as the initial state.

**Calculation.** Figure 2 shows an EB process that relies on the absorption of an incident photon. As we mentioned in Ref. [10] an EB process of this type can be effectively treated as a “generalized” electric dipole transition from the initial to the final state. If the incident radiation with spectral intensity  $I_\omega$  is isotropic and unpolarized, the relation between the probabilities  $W_{ab}$  of a spontaneous transition  $a \rightarrow b$  and  $W_{ba}^{\text{in}}$  of the corre-

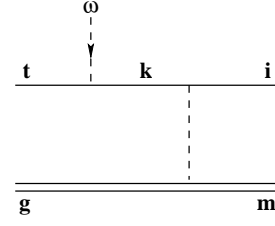


FIG. 2: Simplified Feynman diagram of the considered electronic bridge process. Here it is assumed that the first excitation step leads to efficient population of the state  $(6d7s7p) J = 5/2$  at  $24874 \text{ cm}^{-1}$  which is denoted as  $t$ .

sponding induced transition  $b \rightarrow a$  is given by [11]

$$W_{ba}^{\text{in}} = W_{ab} \frac{4\pi^3 c^2}{\omega^3} I_\omega. \quad (1)$$

The spontaneous EB process can be formally described by the mirror-image of Fig. 2 with an outgoing photon arrow. The general formalism was discussed in detail in two recent articles [7, 10]. Assuming the resonance character of the EB process the expression for  $\Gamma_{\text{EB}}$  can be written as [7]:

$$\Gamma_{\text{EB}} \approx \frac{4}{9} \left( \frac{\omega}{c} \right)^3 \frac{|\langle I_g || \mathcal{M}_1 || I_m \rangle|^2}{(2I_m + 1)(2J_t + 1)} G_2, \quad (2)$$

where  $\mathcal{M}_1$  is the magnetic dipole nuclear moment and  $|I_g\rangle = 5/2^+$  [633] and  $|I_m\rangle = 3/2^+$  [631] are the ground and the isomeric nuclear state, respectively, given in their Nilsson classification.  $J_i$  is the electron total angular momentum of the initial state and  $\omega = \omega_N - \omega_1$  is the frequency of an absorbed photon. The explicit expression for the coefficient  $G_2$  was derived in [10] and reads as

$$G_2 = \sum_{J_k} \frac{1}{2J_k + 1} \times \left| \sum_{\gamma_s} \frac{\langle \gamma_i J_i || \mathcal{T}_1 || \gamma_s J_k \rangle \langle \gamma_s J_k || D || \gamma_t J_t \rangle}{\omega_{si} - \omega_N + i\Gamma_s/2} \right|^2, \quad (3)$$

where  $\omega_{si} \equiv \varepsilon_s - \varepsilon_i$ , and  $\varepsilon_l$  denotes the energy of level  $l$ ,  $\mathcal{T}_1$  is the electronic magnetic-dipole hyperfine coupling operator,  $D$  is the electric dipole moment operator, and  $\gamma_s$  encapsulates all other electronic quantum numbers. The explicit expressions for the matrix elements of the operators  $\mathcal{T}_1$  and  $D$  are given in Ref. [10]. The possible values of the total angular momentum  $J_k$  are determined by the selection rules of the operators  $\mathcal{T}_1$  and  $D$ . In particular, in our case  $J_i = 3/2$  and  $J_t = 5/2$  and, correspondingly,  $J_k$  can be equal to  $3/2$  and  $5/2$ .

In the following we will use the dimensionless quantity  $\beta_{M1}$  introduced in Ref. [10] and defined as the ratio of the probability of the spontaneous EB process  $\Gamma_{\text{EB}}$  to the probability of the spontaneous  $M1$  radiative nuclear  $m \rightarrow g$  transition  $\Gamma_N$ :

$$\beta_{M1} = \frac{\Gamma_{\text{EB}}}{\Gamma_N} \approx \left( 1 - \frac{\omega_1}{\omega_N} \right)^3 \frac{G_2}{3(2J_t + 1)}. \quad (4)$$

We use the method of calculation described in detail in Ref. [7]. We consider  $\text{Th}^+$  as the atom with three valence electrons above the closed-shell core  $[1s^2, \dots, 6p^6]$  and employ the CI+MBPT approach combining the configuration-interaction (CI) method in the valence space with many-body perturbation theory (MBPT) for core polarization effects [12]. At the first stage we solved Dirac-Hartree-Fock (DHF) equations [13] in  $V^{N-3}$  approximation and then we determined the  $5f$ ,  $6d$ ,  $7p$ ,  $7s$ , and  $8s$  orbitals from the frozen-core DHF equations. The virtual orbitals were determined with the help of a recurrent procedure [14]. The one-electron basis set included  $1s$ – $18s$ ,  $2p$ – $17p$ ,  $3d$ – $16d$ , and  $4f$ – $15f$  orbitals on the CI stage.

We formed the configuration spaces allowing single, double, and triple excitations from the  $6d^27s$  configuration (for the even states) and from the  $5f7s^2$  configuration (for the odd states) to the  $7s$ – $13s$ ,  $7p$ – $12p$ ,  $6d$ – $11d$ , and  $5f$ – $10f$  shells. An inclusion of all possible (up to triple) excitations allows us to take into account most completely the configuration interaction for all considered states. The energies and the wave functions are determined from the eigenvalue equation in the model space of the valence electrons

$$H_{\text{eff}}(E_p) |\Phi_p\rangle = E_p |\Phi_p\rangle, \quad (5)$$

where the effective Hamiltonian is defined as

$$H_{\text{eff}}(E) = H_{\text{FC}} + \Sigma(E). \quad (6)$$

Here  $H_{\text{FC}}$  is the relativistic three-electron Hamiltonian in the frozen core approximation and  $\Sigma(E)$  is the energy-dependent core-polarization correction.

Together with the effective Hamiltonian  $H_{\text{eff}}$  we introduce an effective electric-dipole operator  $D_{\text{eff}}$  and an operator  $(\mathcal{T}_1)_{\text{eff}}$  acting in the model space of valence electrons. These operators were obtained within the relativistic random-phase approximation (RPA) [15, 16]. On the stage of solving the RPA equations and calculating diagrams for effective Hamiltonian and effective operators  $D$  and  $\mathcal{T}_1$  we used a more extended basis set. It consisted of  $1s$ – $22s$ ,  $2p$ – $22p$ ,  $3d$ – $22d$ ,  $4f$ – $22f$ , and  $5g$ – $16g$  orbitals.

**Results and discussion.** In the following, we assume that the value of  $\omega_N$  is between 60 000 and 64 000  $\text{cm}^{-1}$ , which corresponds to the range 7.4–7.9 eV and suppose that the main contribution to  $G_2$  (see Eq. (3)) comes from intermediate states lying in this range. As noted above, the odd-parity state  $(6d7s7p) J = 5/2$  at 24874  $\text{cm}^{-1}$  is assumed as the initial state  $t$ . In [10] we discussed that the largest value of the coefficient  $\beta_{M1}$  is expected if the initial state and the intermediate state  $k$  (see Fig. 2) are connected by an  $E1$  transition. Thus, the states  $t$  and  $k$  should be of opposite parity and we have to consider the transition  $(6d7s7p, J = 5/2) \xrightarrow{E1} k \xrightarrow{\mathcal{T}_1} (6d^27s, J = 3/2)$ .

Using Eq. (3) we obtain

$$G_2 \approx \frac{1}{4} \sum_{\gamma_s} \frac{R_{s,J_k=3/2}}{(\omega_{si} - \omega_N + i\Gamma_s/2)^2} + \frac{1}{6} \sum_{\gamma_p} \frac{R_{p,J_k=5/2}}{(\omega_{pi} - \omega_N + i\Gamma_p/2)^2}, \quad (7)$$

where the quantity  $R_{s,J_k}$  is determined as

$$R_{s,J_k} \equiv |\langle 6d^27s, J = 3/2 | \mathcal{T}_1 | \gamma_s J_k \rangle \times \langle \gamma_s J_k | D | 6d7s7p, J = 5/2 \rangle|^2. \quad (8)$$

As follows from Eq. (4) the equation for  $\beta_{M1}$  reads as

$$\beta_{M1} \approx \frac{1}{18} \left( 1 - \frac{\omega_1}{\omega_N} \right)^3 G_2 \approx 0.012 \times G_2, \quad (9)$$

where we took into account that the quantity  $\omega_1/\omega_N \simeq 0.4$  if  $\omega_N$  is between 60000 and 64000  $\text{cm}^{-1}$ .

High-lying energy levels of  $\text{Th}^+$  in the range from 60000 to 64000  $\text{cm}^{-1}$  were determined theoretically in [7]. In Table I we list the values of the coefficients  $R_{s,J_k}$  found for the even-parity states in the frame of the CI+MBPT+RPA approximation. The good correspondence between experimental and theoretical levels at lower energies makes us confident that the level structure is complete for the considered electron configurations. We expect that there will be more levels from other configurations but that the coefficients  $R$  for configurations with multiple electron excitations are smaller than the dominant ones calculated here.

Using Eq. (7), the tabulated coefficients  $R_{s,J_k}$ , and energy values we can find  $G_2$  and  $\beta_{M1}$  for a given value of  $\omega_N$ . The values of  $G_2$  and  $\beta_{M1}$  depend critically on the position of  $\omega_N$  relative to the electronic levels. This is indicated in Fig. 3 which shows the variation of  $G_2$  with  $\omega_N$  in the considered wavenumber range. As seen from this figure,  $G_2 \geq 800$  if  $\omega_N$  lies within the range of the electronic energy levels listed in Table I. For  $\omega_N = 7.6$  eV [5], the  $(5f6d7p) J = 5/2$  state at the calculated value 60462  $\text{cm}^{-1}$  yields the main contribution to the EB process and one obtains  $G_2 \sim 2700$  and  $\beta_{M1} \sim 30$ . When  $G_2$  can be calculated on the basis of spectroscopically determined energy levels, it is likely that the dependence of  $G_2$  on  $\omega_N$  will look somewhat different from Fig. 3. Nevertheless, we expect that the EB excitation rate depends on a small number of dominant channels or even only a single one and that the typical minimum value of the coefficient  $G_2$  will remain at the same order of magnitude ( $\sim 10^3$ ).

The excitation probability to the isomeric state due to resonant laser excitation with  $\omega_1 + \omega = \omega_N$  can be estimated as follows. We assume that radiation at  $\omega$  is produced by a laser source which emits pulses with 10 mJ energy and with a spectral width of  $\Delta\omega = 2\pi \times 3$  GHz at a repetition rate of 30 Hz. These characteristics can be achieved, for example, with commercially available

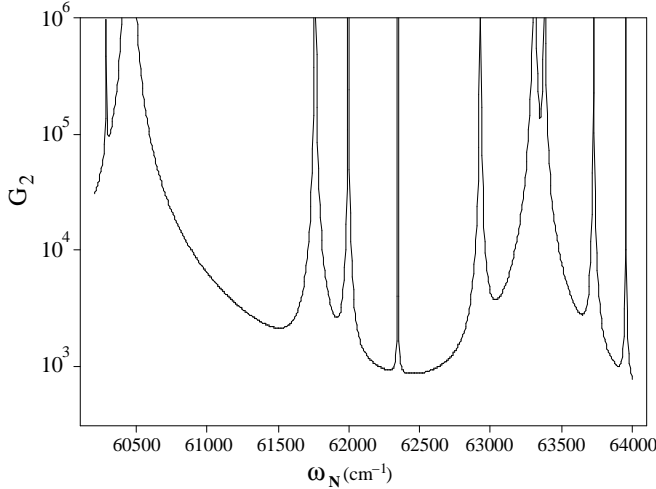


FIG. 3: Dependence of the coefficient  $G_2$  (see Eq. (7)) on the nuclear transition frequency  $\omega_N$  as calculated on the basis of the data listed in Table I.

TABLE I: Calculated energy levels (see text) in the range from 60000 to 64000  $\text{cm}^{-1}$  and coefficients  $R_{k,J_k}$ .  $\Delta_k$  is the difference between the energies of the excited state and the ground state. The notation  $y[x]$  means  $y \times 10^x$ .

$k$	$J_k$	$\Delta_k^a$ ( $\text{cm}^{-1}$ )	$R_{k,J_k}$ (a.u.)
5f6d7p	3/2	60287	3[-4]
6d7s8s	5/2	60416	3[-2]
5f6d7p	5/2	60462	2[-1]
6d7s8s	3/2	61763	2[-3]
5f <sup>2</sup> 6d	5/2	61996	5[-4]
6d <sup>2</sup> 8s	5/2	62345	1[-5]
6d7s8s	3/2	62927	9[-4]
6d <sup>2</sup> 7d	5/2	63308	2[-2]
6d <sup>2</sup> 7d	3/2	63381	4[-3]
6d <sup>2</sup> 7d	3/2	63729	3[-4]
6d <sup>2</sup> 8s + 6d7s8s	5/2	63955	3[-5]

<sup>a</sup>Reference [7].

frequency-doubled dye lasers as they are used in many laboratory applications. With focussing to a beam cross section of  $0.1 \times 0.1 \text{ mm}^2$ , the resulting time-averaged spectral intensity is  $I \approx 1.5 \times 10^{-3} \text{ (W/m}^2\text{) s}$ . Taking into account Eqs. (1) and (4) and assuming  $\beta_{M1} \approx 30$  and the value  $\Gamma_N \simeq 6.6 \times 10^{-4} \text{ s}^{-1}$  [10], we find an excitation rate in the range of  $W_{\text{EB}}^{\text{in}} \approx 10 \text{ s}^{-1}$ . Significantly larger excitation rates can be expected if a suitable electronic transition frequency happens to be very close to  $\omega_N$ .

**Conclusion.** We have suggested a new experimental scheme to excite the nuclear  $^{229\text{g}}\text{Th} - ^{229\text{m}}\text{Th}$  transition in  $\text{Th}^+$  ions and to accurately determine its frequency. The scheme relies on an electronic bridge process that is driven by two incident laser photons whose sum frequency is resonant with the nuclear transition frequency. Using our previous calculations of the electronic energy level structure of  $\text{Th}^+$  in the experimentally relevant energy range [7], we have estimated the probability of the investigated two-photon electron bridge process. Assuming that the nuclear transition energy is close to 7.6 eV, we find that a nuclear excitation probability in the range of  $10 \text{ s}^{-1}$  can be obtained with only moderate laser power and bandwidth requirements.

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- [1] E. Peik and C. Tamm, *Europhys. Lett.* **61**, 181 (2003).
  - [2] V. V. Flambaum, *Phys. Rev. Lett.* **97**, 092502 (2006).
  - [3] L. A. Kroger and C. W. Reich, *Nucl. Phys. A* **259**, 29 (1976).
  - [4] C. W. Reich and R. G. Helmer, *Phys. Rev. Lett.* **64**, 271 (1990).
  - [5] B. R. Beck, J. A. Becker, P. Beiersdorfer, G. V. Brown, K. J. Moody, J. B. Wilhelmy, F. S. Porter, C. A. Kilbourne, and R. L. Kelley, *Phys. Rev. Lett.* **98**, 142501 (2007).
  - [6] E. V. Tkalya, *Phys. Uspekhi* **46**, 315 (2003).
  - [7] S. G. Porsev and V. V. Flambaum, *Phys. Rev. A* **81**, 042516 (2010).
  - [8] URL <http://www.lac.u-psud.fr/Database/Tab-energy/Th161m/TKalya-Wr.Rtml>
  - [9] K. Zimmermann *et al.* (to be published).
  - [10] S. G. Porsev and V. V. Flambaum, *Phys. Rev. A* **81**, 032504 (2010).
  - [11] I. I. Sobelman, *Atomic Spectra And Radiative Transitions* (Springer-Verlag, Berlin, Heidelberg, New York, 1979).
  - [12] V. A. Dzuba, V. V. Flambaum, and M. G. Kozlov, *Phys. Rev. A* **54**, 3948 (1996).
  - [13] V. F. Brattsev, G. B. Deyneka, and I. I. Tupitsyn, *Bull. Acad. Sci. USSR, Phys. Ser. (Engl. Transl.)* **41**, 173 (1977).
  - [14] M. G. Kozlov, S. G. Porsev, and V. V. Flambaum, *J. Phys. B* **29**, 689 (1996).
  - [15] V. A. Dzuba, M. G. Kozlov, S. G. Porsev, and V. V. Flambaum, *Zh. Eksp. Teor. Fiz.* **114**, 1636 (1998), [*Sov. Phys.-JETP* **87** 885, (1998)].
  - [16] J. D. Koelb, W. R. Johnson, and P. Shorer, *Phys. Rev. A* **26**, 19 (1982).